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D. M. Dearborn and S. C. Keeton

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Lawrence Livermore National Laboratory Experience Using 30-Gallon Drum Neutron Multiplicity Counter for Measuring Plutonium-Bearing Salts

David Dearborn, Lawrence Livermore National Laboratory
7000 East Avenue, L-347, Livermore, CA 94550, USA (925) 424-6974
Stewart Keeton, Lawrence Livermore National Laboratory
7000 East Avenue, L-347, Livermore, CA 94550, USA (925) 422-9800

Abstract

Lawrence Livermore National Laboratory (LLNL) has been performing accountability measurements of plutonium (Pu) -bearing items with the 30-gallon drum neutron multiplicity counter (NMC) since August 1998. A previous paper¹ focused on the LLNL experience with Pu-bearing oxide and metal items. This paper expands on the LLNL experience with Pu-bearing salts containing low masses of Pu. All Pu-bearing salts used in this study were measured using calorimetry and gamma isotopic analyses (Cal/Iso) as well as the 30-gallon drum NMC. The Cal/Iso values were treated as being the true measure of Pu content because of the inherent high accuracy of the Cal/Iso technique, even at low masses of Pu, when measured over a sufficient period of time. Unfortunately, the long time period required to achieve high accuracy from Cal/Iso can impact other required accountability measurements. The 30-gallon drum NMC is a much quicker system for making accountability measurements of a Pu-bearing salt and might be a desirable tradeoff. The accuracy of 30-gallon drum NMC measurements of Pu-bearing salts, relative to that of Cal/Iso, is presented in relation to the mass range and alpha associated with each item. Conclusions drawn from the use of the 30-gallon drum NMC for accountability measurements of salts are also included.

Introduction

The Lawrence Livermore National Laboratory (LLNL) 30-gallon drum neutron multiplicity counter (NMC) was designed, built, and initially characterized [with ²⁵²Cf, uranium (U), and plutonium (Pu) standards] at Los Alamos National Laboratory (LANL). It was delivered to LLNL in May 1995 and installed in the LLNL Plutonium Facility in September 1995.

The 30-gallon drum NMC was certified for accountability measurements in July 1997 after completion of an extensive measurement effort involving the Calex 1 weapons-grade Pu oxide standard (nominal 400 g of Pu) and several well-characterized Pu oxide (10 g to 1150 g) and Pu metal (90 g to 2625 g) items from the LLNL inventory. Between July 1997 and October 1999, measurement control of the 30-gallon drum NMC was based solely on the Calex 1 Pu oxide standard. In October 1999, a second measurement control chart was established based on the STDISO12L fuels-grade Pu oxide standard (nominal 25g of Pu).

A previous paper (Reference 1) focused on the LLNL experience with Pu-bearing oxide and metal items. The accuracy (one standard deviation) for both Pu-bearing oxide or metal items found in that study was between 2% and 3%. Most of the Pu-bearing salt items were removed from the databases studied in the previous paper. This was done because either the book mass was a very poor estimate of the actual Pu mass or because the alpha value was larger than an arbitrary upper limit of 5, but more importantly, because salts generally have large masses (3 or more kg) with very small amounts of Pu. Therefore, the Pu mass uncertainties are generally

larger than the nominal upper limit of 5% observed from other kinds of Pu-bearing items. In addition, the preliminary results found in that study appeared to show that high alpha values for salts did not have much influence on the accuracy of the Pu mass content from 30-gallon drum NMC measurements. Furthermore, when the salts were studied as a group, they appeared to give statistically valid results with a one standard deviation accuracy of about 25% for Pu masses less than about 40 g.

This paper describes additional LLNL experience with Pu-bearing salts (referred to herein as salts) containing less than 400 grams of Pu. All salts used in this study were measured using both calorimetry and gamma isotopic analyses (Cal/Iso), in addition to using NMC. The Cal/Iso values are treated as being the actual Pu-mass values because of the inherent high accuracy of Cal/Iso even at low masses of Pu when measured over a sufficient time span. However, the long time necessary to get high accuracy from Cal/Iso can impact other required MC&A measurements. NMC measurements generally require 20 minutes or so, as compared to six or more hours for calorimetry. The NMC measurements are therefore much quicker and can be considered a desirable tradeoff. The accuracy of NMC measurements for salts, relative to that of Cal/Iso, will be presented by the mass range and alpha value associated with each one. Conclusions drawn from the use of the 30-gallon drum NMC on salts are included.

Results

As mentioned, salt items generally have large masses (3 or more kg) with very small amounts of Pu. One consequence of this is that the presence of large amounts of unknown material with potentially substantial (α, n) reactions can give rise to large alpha values. In addition, the book mass assigned during chemical processing is generally a very poor estimate of the actual Pu mass. This is because chemical separation processes frequently produce unexpected residual amounts of Pu in salts that can be significantly more than or less than the projected amounts. Hence the projected book value cannot be used as the known Pu mass. Therefore, a Cal/Iso measured mass was chosen as a good value for the unknown Pu mass.

The actual Pu mass found in salt items can be placed into three different mass ranges. The mass ranges chosen are 0-10 g, 10-40 g and 40-400 g of Pu. The 0-10 g mass range represents the expected mass range from successful chemical separation processes. When the chemical separation process does not succeed quite as well as desired, the salt items will have Pu masses in the 10-40 g mass range. While a failed chemical separation run can also yield salts having Pu masses in the 40-400 g mass range. Failed separation runs can yield salt items with Pu mass greater than 400 g. Pu mass values greater than 400 g are considered as being impure materials and are not lumped with the salts, as this implies an even more severe failure in the chemical separation process.

The results for each 0-10 g salt, shown graphically in Figure 1, shows the cumulative number of residuals vs. residual (difference between Cal/Iso and NMC/Iso divided by the standard deviation), and are summarized in Table 1. Also shown in the figure is the cumulative distribution curve that the results of a normally distributed process would follow. Data from 0-10 g salts with alpha less than -4 or greater than 200 were excluded from this study since they appeared to prejudice the results, however, the choice of these values is somewhat arbitrary. Notice that the residuals are distributed around this curve, and this means that the data points do appear to be normally distributed in a statistical sense (exact normal distribution would mean all data points would lie on the curve). The corresponding alpha values matching each data point are

also displayed in Figure 1. The values displayed for alpha are divided by 100 so the same ordinate can be used. Notice that there seems to be essentially no correlation between the alpha value and the residual value, although the smallest ones do seem to be mostly associated with negative residuals. The largest value for alpha on the graph is 82 and the smallest one is -1.2. The value derived for the standard deviation from the data in Figure 1 is 24% with a bias of -2.3%.

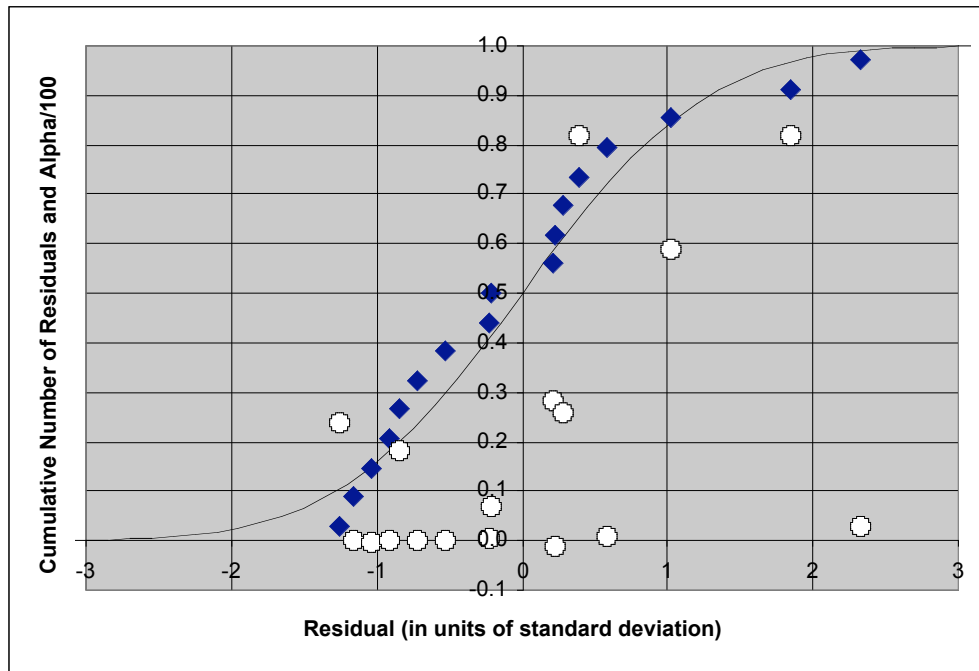


Figure 1: shows the cumulative number of residuals vs. residual for 0-10 g salts (diamonds), and the corresponding alpha values divided by 100 (open circles). The smooth curve gives the cumulative normal distribution curve.

Table 1 shows a summary of the results for all salts where both NMC/Iso and Cal/Iso measurements have been obtained for the three mass range categories mentioned above. LLNL has measured many more salt items in the 30-gallon drum NMC than have also been measured in Cal/Iso. This is because the measurement equipment is used primarily for mass measurements of Pu-bearing materials in support of plutonium technology programs at LLNL, and not for use in an R&D measurement effort.

Table 1: Standard deviation and bias for the Pu-bearing salt items where both NMC/Iso and Cal/Iso Pu-mass values were determined. These salt values are separated into the three mass ranges designated. The range of alpha values from the figures is also given.

Mass range (g)	Number of Items	Minimum Alpha Value	Maximum Alpha Value	Standard Deviation (%)	Bias (%)
0-10	17	-1.2	81.8	24	-2.3
10-40	21	-3.8	29.1	22	1.8
40-400	16	0.4	13.3	17	-1.4

Figure 2 shows the cumulative number of residuals vs. residual for each 10-40 g salt. Also shown in the figure is the cumulative normal distribution curve. Data from 10-40 g salts with alpha greater than 100 were excluded from this study since they appeared to prejudice the results (again a somewhat arbitrary choice). Notice that the residuals shown here appear to be normally distributed in a statistical sense, as they are even closer to the cumulative normal distribution curve than are those for 0-10 g salts. The alpha value corresponding to each data point is also displayed in Figure 2. Again the alpha values are divided by 100. There seems to be no correlation between the alpha value and the residual value. The largest alpha value on the graph is 29 and the smallest one is -3.8 . The standard deviation derived from the data in Figure 2 is 22% with a bias of 1.8%.

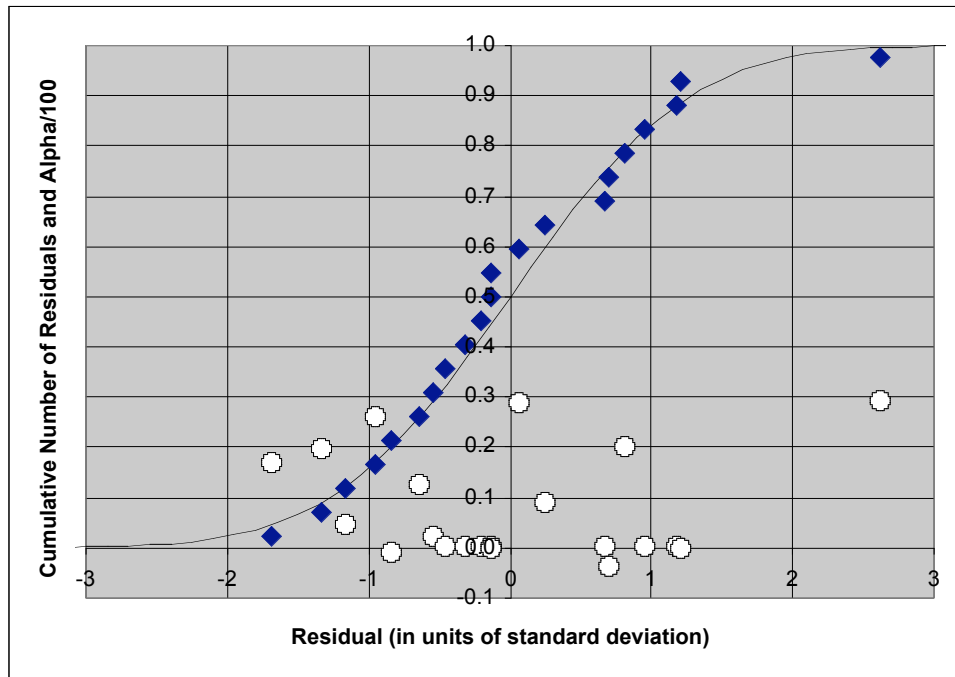


Figure 2: shows the cumulative number of residuals vs. residual for 10-40 g salts (diamonds), and the corresponding alpha values divided by 100 (open circles). The smooth curve gives the cumulative normal distribution curve.

Figure 3 shows the cumulative number of residuals vs. residual for each 40-400 g salt. Also shown in the figure is the cumulative normal distribution curve. The residuals here appear to be approximately scattered in a normal distribution manner. The alpha value corresponding to each data point is also displayed in Figure 2. Again the alpha values are divided by 100. Notice that there seems to be no correlation between the alpha value and the residual value. The largest alpha value on the graph is 13 and the smallest one is 0.4. The value derived for the standard deviation from the data in Figure 2 is 17% with a bias of -1.4% .

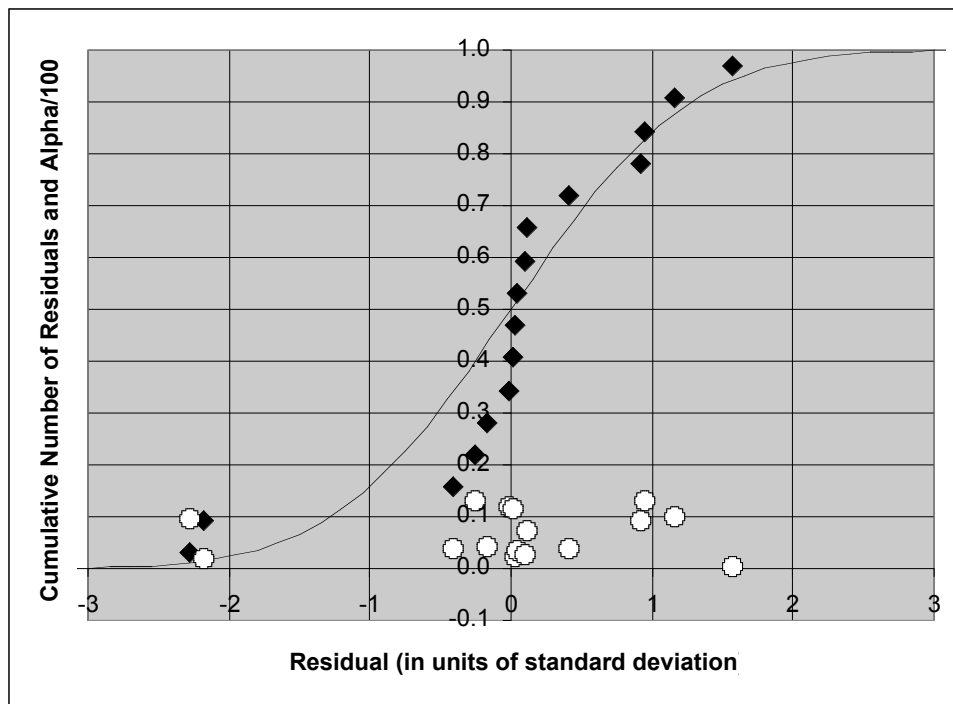


Figure 3: shows the cumulative number of residuals vs. residual for 40-400 g salts (diamonds), and the corresponding alpha values divided by 100 (open circles). The smooth curve gives the cumulative normal distribution curve.

Conclusions and Recommendations

The standard deviation and bias for the salts given in Table 1 and the three figures indicate that there is only a small bias for the data compared to a normal distribution. This lends credence to the proposal that the data points are normally distributed. For 0-10 g salts we found that values of alpha as large as about 100 or so do not appear to have a significant effect on the accuracy of the NMC/Iso measured mass values for a salt in this mass range. For salts with Pu masses greater than 10 g we found that alpha values up to about 30 do not appear to have a significant effect on NMC accuracy. A negative alpha value of less than about -4 does seem to affect this accuracy, however. The 30-gallon drum NMC is a valuable tool when measuring salts containing low Pu mass. When faced with measuring a large number of salts in a short time period, the 30-gallon drum NMC is the system of choice.

Acknowledgments

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References

- ¹ D. Dearborn and M. Mount, "Lawrence Livermore National Laboratory Experience with the 30-Gallon Drum Neutron Multiplicity Counter," in Proceedings of the 44th Annual Meeting, Institute of Nuclear Materials Management, Phoenix, AZ, July 13-17, 2003.

University of California
Lawrence Livermore National Laboratory
Technical Information Department
Livermore, CA 94551

